# Molecular dynamics algorithms for quantum Monte Carlo methods



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**Abstract**

In the present Letter, novel molecular dynamics methods compatible with corresponding quantum Monte Carlo methods are developed. One is a variational molecular dynamics method that is a molecular dynamics analog of quantum variational Monte Carlo method. The other is a variational path integral molecular dynamics method, which is based on the path integral molecular dynamics method for finite temperature systems by Tuckerman *et al.*, J. Chem. Phys. **99**, 2796 (1993). These methods are applied to model systems including the liquid helium-4, demonstrated to work satisfactorily for the tested ground state calculations.

## 1. Introduction

Quantum Monte Carlo (QMC) methods have recently attracted great interest in the field of computational physics and chemistry as tools for accurately calculating ground state properties of many body systems [1, 2, 3, 4]. Variational Monte Carlo (VMC) method [5], for example, is used to calculate

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expectation values of physical quantities using a trial wavefunction of the system. The more sophisticated diffusion Monte Carlo (DMC) method [6, 7] is a projector approach in which a stochastic imaginary time evolution is used to improve a starting trial wavefunction. The QMC methods including the VMC and DMC methods have successfully been applied to various quantum systems ranging from quantum liquids like helium to electronic structure of atoms and molecules [1, 4]. In the present study, we focus on the variational Monte Carlo and a variational path integral [1, 8] methods; the latter is closely related to the diffusion Monte Carlo method.

In this Letter, we develop novel molecular dynamics methods for two quantum Monte Carlo methods. One is a variational molecular dynamics (VMD) method, which is a molecular dynamics analog of the variational Monte Carlo method. Square modulus of a trial wavefunction used in the VMC method is reinterpreted to be a canonical distribution at a fictitious temperature. Then, a molecular dynamics method is constructed using an extended system method like a Nosé-Hoover chain thermostat [9] to generate the fictitious canonical distribution. The other is a molecular dynamics method for the variational path integral method [1]. The variational path integral, which is also called path integral ground state [8], is a method to numerically generate exact ground state of many body systems. In the present study, we construct a molecular dynamics algorithm to carry out the variational path integral calculations on the basis of path integral molecular dynamics method developed for finite temperature quantum systems [10, 11]. We call it a variational path integral molecular dynamics (V-PIMD) method. Then, we apply the VMD and V-PIMD methods to model systems for show-

ing reliability of the present algorithms.

## 2. Methodology

### *2.1. Variational molecular dynamics*

We start to consider a system consisting of $N$ particles whose coordinates are collectively represented to be $R$. The ground state of the system is described using a trial wavefunction $\Phi_T(R, \alpha)$ where $\alpha$ denotes a set of variational parameters. Expectation value of a hamiltonian $\hat{H}$ using the trial wavefunction reads [1, 4]

$$\begin{aligned} \langle H \rangle &= \frac{\int dR \Phi_T^*(R) \hat{H} \Phi_T(R)}{\int dR |\Phi_T(R)|^2} \\ &= \frac{\int dR |\Phi_T(R)|^2 E_L(R)}{\int dR |\Phi_T(R)|^2}, \end{aligned} \tag{1}$$

where $E_L(R)$ represents a local energy defined by

$$E_L(R) = \frac{\hat{H}\Phi_T(R)}{\Phi_T(R)}. \tag{2}$$

The variational Monte Carlo method is designed so as to generate coordinates $R$ according to a distribution function $\rho(R) \propto |\Phi_T(R)|^2$. Then, the expectation value of the hamiltonian Eq. (1) can be calculated by the local energy averaged along a Monte Carlo trajectory. Here, we consider a molecular dynamics method to generate the distribution function $\rho(R)$. First, we define an effective potential $V_{\mathrm{VMC}}(R)$ using the distribution function $\rho(R)$:

$$\rho(R) \equiv e^{-\beta V_{\mathrm{VMC}}(R)}, \tag{3}$$

where a parameter $\beta$ is a fictitious inverse temperature. The distribution function $\rho(R)$ can be regarded as a canonical distribution at the fictitious $\beta$.

To sample the distribution, we introduce the following classical hamiltonian:

$$H_{\mathrm{VMC}} = \sum_{i=1}^{N} \frac{\mathbf{p}_i^2}{2m_i'} + V_{\mathrm{VMC}}(R), \tag{4}$$

where $\mathbf{p}_i$ is a fictitious momentum of an $i$-th particle and $m_i'$ is an associated fictitious mass. Then, we can obtain equations of motion based on the Hamilton equation. Furthermore, we attach a single Nosé-Hoover chain thermostat to the system for generating the canonical distribution Eq. (3). The resulting thermostatted equations of motion are basic equations of the variational molecular dynamics method. Then, as in the variational Monte Carlo method, we can obtain quantum mechanical expectation values of various physical quantities using a molecular dynamics trajectory.

*2.2. Variational path integral molecular dynamics*

In this subsection, a method to systematically improve the description of the system is considered. The exact ground state of the system, $|\Psi_0\rangle$, can be obtained using a trial wavefunction $|\Phi_T\rangle$ by the following relation: [1, 4]

$$|\Psi_0\rangle = \lim_{\beta\to\infty} e^{-\frac{\beta}{2}\hat{H}}|\Phi_T\rangle, \tag{5}$$

where $\beta$ is an imaginary time [1]. Here, we refer to a scalar product of the above exact wavefunction as a pseudo partition function $Z_0$ [12], which plays a central role to construct a variational path integral:

$$\begin{aligned} Z_0 &= \langle\Psi_0|\Psi_0\rangle = \langle\Phi_T|e^{-\beta\hat{H}}|\Phi_T\rangle \\ &= \int\int dRdR'\langle\Phi_T|R\rangle\langle R|e^{-\beta\hat{H}}|R'\rangle\langle R'|\Phi_T\rangle, \end{aligned} \tag{6}$$

where we use the closure relation for the coordinate basis: $\int dR|R\rangle\langle R| = 1$. A matrix element $\langle R|e^{-\beta\hat{H}}|R'\rangle$ in Eq. (6) is found to be the same as a density

matrix at the inverse temperature $\beta$, $\rho(R, R'; \beta)$. The density matrix can be written on the basis of the discretized path integral as [1]

$$\begin{aligned} \rho(R, R'; \beta) &= \langle R|[e^{-\Delta\tau\hat{H}}]^M|R'\rangle \\ &= \int\cdots\int\{\prod_{s=1}^{M-1} dR^{(s)}\}\prod_{s=0}^{M-1}\langle R^{(s)}|e^{-\Delta\tau\hat{H}}|R^{(s+1)}\rangle \\ &\propto \int\cdots\int\{\prod_{s=1}^{M-1} dR^{(s)}\}e^{-S(\{R^{(s)}\};\Delta\tau)}, \end{aligned} \tag{7}$$

where $\Delta\tau = \beta/M$ and $S(\{R^{(s)}\}; \Delta\tau)$ is a discretized imaginary time action. Explicit expression of the action is dependent on an approximation on a short time propagator $\langle R|e^{-\Delta\tau\hat{H}}|R'\rangle$. In the present study, the primitive approximation [1] is adopted. Then, the pseudo partition function can be written by

$$Z_0 \propto \int\cdots\int\{\prod_{s=0}^{M} dR^{(s)}\}\Phi_T(R^{(0)})e^{-S(\{R^{(s)}\};\Delta\tau)}\Phi_T(R^{(M)}). \tag{8}$$

As in the standard path integral method for finite temperature systems [13], the pseudo partition function can be regarded as a configurational integral of classical polymers. However, in the variational path integral, the classical isomorphic systems consist of open chain polymers. Furthermore, distributions of end-point coordinates at $s = 0$ and $M$ are affected by the trial wavefunction $\Phi_T(R^{(0)})$ and $\Phi_T(R^{(M)})$, respectively. Here, we consider a molecular dynamics method to sample configurations of the above isomorphic polymers. First, we define the following classical hamiltonian:

$$H_{\mathrm{V-PIMD}} = \sum_{s=0}^{M}\sum_{i=1}^{N}\frac{(\mathbf{p}_i^{(s)})^2}{2m_i'} + \frac{S(\{R^{(s)}\})}{\beta} - \frac{\ln\Phi_T(R^{(0)})}{\beta} - \frac{\ln\Phi_T(R^{(M)})}{\beta}, \tag{9}$$

where $\mathbf{p}_i^{(s)}$ denotes a fictitious momentum of an $i$-th particle at an $s$-th time slice and $m_i'$ is a fictitious mass of the $i$-th particle. Using the above hamiltonian, we can derive equations of motion based on the Hamilton equation. Then, to generate the distribution compatible with Eq. (8), we attach a single Nosé-Hoover chain thermostat to each degree of freedom. The resulting equations of motion are basic equations for the variational path integral molecular dynamics (V-PIMD) method. In the present study, we use staging coordinates [10] to describe the polymer configurations for enhancing sampling efficiency. The standard definition [11, 14] on the staging variables and associated staging masses $m^{(s)}$ are adopted.

It is commented that in Eq. (9), the logarithm of the trial wavefunction is needed to define $H_{\mathrm{V-PIMD}}$. However, trial wavefunctions can not be positive everywhere, for example, for fermionic wavefunctions. Although a formally exact formulation is possible using the absolute value of the wavefunction, this scheme suffers from the so-called "sign problem" that is a problem common to projector Monte Carlo methods such as the diffusion Monte Carlo [4]. Thus, one usually resort to the fixed node approximation [4, 7]; it gives ground state energies that satisfy a variational principle and are usually very accurate, which is used in almost all current large scale applications of the diffusion Monte Carlo method [4]. Since in the fixed node approximation, a part of configuration space with the same sign of the trial wavefunction is sampled, this approximation may provide a practical route to construct the variational path integral molecular dynamics method; however, numerical studies are needed to show that the molecular dynamics calculations can be performed stably.

## 3. Results on pilot calculations

### *3.1. Harmonic oscillator*

We first consider a one-dimensional harmonic oscillator as a model system. The hamiltonian of the oscillator is written as

$$\hat{H} = -\frac{\hbar^2}{2m}\frac{d^2}{dx^2} + \frac{1}{2}m\omega^2 x^2. \tag{10}$$

Hereafter, we use units by which $m = \omega = \hbar = 1$. To describe the ground state of the oscillator, the following trial wavefunction is employed:

$$\Phi_T(x) \propto e^{-\alpha x^2}, \tag{11}$$

where $\alpha$ is a variational parameter. When the parameter $\alpha = 0.5$, the trial wavefunction becomes exact. The expectation value of the hamiltonian with the trial wavefunction can be calculated analytically as

$$\langle \hat{H} \rangle = \frac{1}{2}\left(\alpha + \frac{1}{4\alpha}\right). \tag{12}$$

On the other hand, the local energy Eq. (2) for the above trial wavefunction has the following form:

$$E_L(x) = \alpha + \left(\frac{1}{2} - 2\alpha^2\right) x^2. \tag{13}$$

Here, we summarize computational details on the oscillator calculations. For the variational molecular dynamics (VMD) calculations, the fictitious inverse temperature $\beta$ was chosen to be 1.0 and the fictitious mass was set to be equal to the oscillator's mass: $m' = m = 1.0$. The equations of motion were integrated with a time step $\Delta t = 2\pi/100$. The VMD calculations have been performed for various values of the variational parameter $\alpha$. For each

$\alpha$, the VMD calculation was performed 500000 steps. On the other hand, for the variational PIMD, the imaginary time increment $\Delta\tau$ was chosen to be 0.1 and the number of time slices $M = 100$, and then the total imaginary time $\beta = 10.0$. As noted in Sec. 2.2, the V-PIMD calculation was performed using the staging variables. The fictitious masses for the staging variables $m^{(s)'}$ were set to be equal to the corresponding staging masses except end-point coordinates (at $s = 0$ and $M$) where $m^{(0)'} = m^{(M)'} = \gamma m$. The parameter $\gamma = 0.01$ was used. The V-PIMD calculation was performed 500000 steps with a time increment $\Delta t = 2\pi/(100\omega_M)$ where $\omega_M = \sqrt{M}/\beta$.

In Fig. 1, we show the total energy of the oscillator for various $\alpha$ calculated by the VMD method. It is found that the VMD results are in good agreement with the analytical results for all the cases presented. Since the trial wavefunction becomes exact at $\alpha = 0.5$, the total energy has a minimum at the parameter value. We can find the variance of the energy decreases with approaching the exact quantum state, and the variance vanishes at $\alpha = 0.5$. This corresponds to the well known zero variance property [1, 4] of the variational Monte Carlo method. We next show the averaged potential energy as a function of the imaginary time $\tau$ obtained by the V-PIMD method. The variational parameter of the wavefunction at $s = 0$ and $M$ was chosen to be $\alpha = 0.7$ for showing how the poor description of the ground state is improved by the variational path integral. As seen in Fig. 2, starting from an initial potential energy at $\tau = 0$, the energy quickly decreases and reaches a plateau around $\tau = 2.0$ where the energy is the exact ground state value. Thus, in the time range $\tau = 2.0 \sim 8.0$, the system is in the exact ground state. Actually, the total energy was calculated to be $0.500 \pm 0.005$ using the

mixed estimator derived by the following relation [4]:

$$E_0 = \frac{\langle \Phi_T | \hat{H} e^{-\beta \hat{H}} | \Phi_T \rangle}{\langle \Phi_T | e^{-\beta \hat{H}} | \Phi_T \rangle}, \tag{14}$$

where $E_0$ is the ground state energy. The variational PIMD energy is found to be in perfect agreement with the exact ground state energy. The following probability distribution of the oscillator is also presented in Fig. 3:

$$P(x) = \frac{|\Psi_0(x)|^2}{\int dx |\Psi_0(x)|^2}. \tag{15}$$

While the probability distribution using the trial wavefunction with $\alpha = 0.7$ shows poor description of the system, the V-PIMD is found to give the numerically exact distribution of the oscillator.

Here, we comment on how large $\beta$ one need to obtain the exact ground state when a good trial wavefunction is not available. For an enough large $\beta$, Eq. (14) can be written by

$$\begin{aligned} \frac{\langle \Phi_T | \hat{H} e^{-\beta \hat{H}} | \Phi_T \rangle}{\langle \Phi_T | e^{-\beta \hat{H}} | \Phi_T \rangle} &= -\frac{d}{d\beta} \ln \langle \Phi_T | e^{-\beta \hat{H}} | \Phi_T \rangle \\ &= E_0 + \mathcal{O}(e^{-\beta \Delta E_1}), \end{aligned} \tag{16}$$

where $\Delta E_1$ is the energy difference between the first excited state and the ground state. The above relation can be verified using the trial wavefunction expanded by eigenfunctions of the hamiltonian $\hat{H}$. Using the above relation, $\beta \approx 1/\Delta E_1$ is needed to relax to the exact ground state. In the case of the harmonic oscillator example, $\beta \approx 1$ since $\Delta E_1 = \hbar\omega = 1$ in the units employed.

### *3.2. Liquid helium-4*

Next, we consider the liquid helium-4 at the ground state as a realistic application. To describe the liquid state of the helium-4, the following trial

wavefunction is employed:

$$\Phi_T(R) = \prod_{i<j}^{N} e^{-\frac{1}{2}(\frac{b}{r_{ij}})^5}, \tag{17}$$

where $b$ is a variational parameter. In the present study, $b = 3.07$ Å was adopted for both the VMD and V-PIMD calculations [8]. The density of the system was set to be the experimental equilibrium value $\rho = 0.0218$ Å$^{-3}$. The system consisted of $N = 108$ helium atoms in a cubic box under the periodic boundary condition. The interatomic interaction was represented by the pair interaction developed by Aziz and coworkers [15]. For the VMD calculation, the fictitious inverse temperature was chosen to be 1.0 and the fictitious mass was set to be the physical mass of the helium-4 atom. The VMD calculation was carried out 300000 steps with a time increment $\Delta t = 20$ fs after an equilibration period. On the other hand, for the V-PIMD calculation, the imaginary time increment was set to be $\Delta\tau = 0.001$ K$^{-1}$ and $M = 400$. Then, the total imaginary time was $\beta = 0.4$ K$^{-1}$. The fictitious masses were chosen as in the case of the oscillator in Sec. 3.1 with $\gamma = 0.02$. The V-PIMD calculation was performed 500000 steps with a time increment $\Delta t = 10$ fs.

Figure 4 shows the potential energy of the helium per atom as a function of the imaginary time $\tau$ by the V-PIMD calculation. After an initial transient regime, the potential energy is found to relax to a plateau value around $\tau = 0.1$ K$^{-1}$ where the energy can be expected to be the exact ground-state potential energy for the given hamiltonian. Then, in the imaginary time interval $\tau = 0.1 \sim 0.3$ K$^{-1}$, the system is in the ground state of the helium-4. Using the mixed estimator, the total energy was calculated to be $-7.33 \pm 0.01$ K, which was in good agreement with the experimental value $-7.17$

K [16]. To see atomistic structural correlation in the ground state, the radial distribution function $g(r)$ is presented in Fig. 5. The radial distribution function is defined using the ground state wavefunction as follows:

$$g(r_{12}) = \frac{N(N-1)}{\rho^2} \frac{\int d\mathbf{r}_3 \cdots d\mathbf{r}_N |\Psi_0(\mathbf{r}_1, \ldots, \mathbf{r}_N)|^2}{\int d\mathbf{r}_1 \cdots d\mathbf{r}_N |\Psi_0(\mathbf{r}_1, \ldots, \mathbf{r}_N)|^2}. \tag{18}$$

As shown in the figure, the $g(r)$ by the V-PIMD calculation is more structured than the VMD counterpart. The V-PIMD result is found to be in good agreement with the experimental $g(r)$ [17], indicating the numerical improvement of the trial wavefunction by the variational path integral.

## 4. Discussion

In this section, we compare the present methods with other related quantum Monte Carlo methods. We first discuss the variational molecular dynamics method. For comparison, we have performed a set of the variational Monte Carlo calculations for the liquid helium. The conventional Metropolis [5, 18] and smart Monte Carlo [19, 20] methods have been implemented; in the latter method, the force on a selected atom is used to enhance sampling efficiency in configuration space. In one MC step, we performed $N$ attempts to displace atoms; each VMC run consisted of 300000 MC steps. We also performed a VMD calculation of the liquid helium for 300000 steps with $\Delta t = 40.0$ fs. Using this time increment, the average error of the energy per step was evaluated to be about 0.02 % in the total energy. The mean square displacement of a tagged atom was calculated as a function of MD or MC step; this quantity provides a measure to see sampling efficiency in configuration space. A self-diffusion coefficient $D$ is defined by the slope of the

mean squre displacement. In Fig. 6, the calculated diffusion coefficients by the VMC and VMD methods are presented. Using the standard Metropolis method, the diffusion coefficient becomes large with decreasing the acceptance ratio $P_{acc}$. In the case of $P_{acc} = 31\%$, the diffusion coefficient is almost the same as that by the VMD calculation. The largest $D$ presented in the Figure is provided in the case of $P_{acc} = 15\%$, which is 1.3 times larger than the VMD result. In this case, the standard MC method is 1.3 times more efficient than the VMD method. On the other hand, the diffusion coefficient as a function of the acceptance ratio has a maximum around $P_{acc} = 50\%$ by the smart MC method. In this case, the smart MC method is 1.2 times more efficient than the VMD method. Here, we present the CPU cost needed for the VMC and VMD calculations. The averaged amounts of the CPU time for 300000 steps were 500, 659, and 192 seconds for the standard MC, smart MC, and VMD methods, respectively, using a Xeon 3.0 GHz workstation. The present VMC codes were written using the part of the potential and force calculations in the VMD code. In the present implementation of the MC methods, the potential between a selected atom and other atoms, and the force on a selected atom for the smart MC, were calculated for old and suggested configurations in each trial move, straightforwardly. The VMC and VMD codes were compiled by an intel C compiler without optimization options. We define the following factor to discuss the computational cost: $\zeta = t_{\rm VMC}/t_{\rm VMD}$ where $t_{\rm VMC}$ and $t_{\rm VMD}$ represent the amount of CPU time needed for the VMC and VMD calculations; $\zeta = 2.6$ and 3.4 for the standard and smart MC methods, respectively. We also define an effective diffusion coefficient $D^\star = D/\zeta$ which measures computational efficiency of

the VMC method compared with the VMD method. Owing to the factor $\zeta$, the effective diffusion coefficient by the VMD method ($\zeta = 1$) is larger than those by the VMC methods, although the value of $\zeta$ depends on the details of the codes. These results indicate that the computational efficiency of the VMD method can be competitive with that of the VMC method for the present system.

Here, we move on to the variational path integral molecular dynamics method. The V-PIMD method is constructed on the basis of the path integral molecular dynamics (PIMD) method developed for finite temperature systems. Tuckerman *et al.* [10] have shown that the PIMD method can be almost as efficient as the path integral Monte Carlo method. Then, the efficiency of the V-PIMD method is likely to be competitive with the variational path integral sampled by the Monte Carlo method [1, 8], although direct numerical comparison may be required for quantitative discussion. Compared with the diffusion Monte Carlo (DMC) by which the ground state energy can be calculated accurately, the variational path integral has the advantage of providing relatively easily expectation values for physical quantities that do not commute with the hamiltonian [8]. Although the variational path integral needs a trial wavefunction $\Phi_T$ as in DMC, it has been demonstrated [8] that results for quantities other than the energy are considerably less sensitive to the choice of $\Phi_T$ than in DMC.

Finally, the advantage of the molecular dynamics method over the Monte Carlo method is summarized. In MD, unlike the standard MC method, all the coordinates are simultaneously updated; this feature enables us to use efficient parallel computation algorithms developed for the classical MD [21],

which are useful to perform large scale quantum simulations. Moreover, in V-PIMD, the force calculations at each time slice can almost be performed independently. Thus, the V-PIMD method can efficiently be implemented in parallel computations. Although the same parallel computation procedures can be used in MC, a sequence of local MC moves corresponding to one MD step is difficult to perform in parallel. It is noted that an efficient global update of the coordinates is possible by a Monte Carlo method called the hybrid Monte Carlo (HMC) [10, 22]; this is a combined method of MD and MC. In HMC, trial configurations are generated by equations of motion as in MD. The trial configurations are accepted according to an appropriate Metropolis criterion by which the errors arising from the finite time step $\Delta t$ are eliminated. In HMC, one can use a time step $\Delta t$ which is too long for MD. However, one can not make the time step for a single HMC move too long, because the acceptance would become very small. As a consequence, the performance of HMC is not dramatically better than that of the corresponding MD. Moreover, the acceptance probability of HMC moves with a fixed $\Delta t$ decreases with the systems size, because the root mean square error in the energy increases with $\sqrt{N}$ [18]; on the other hand, the stability of the MD algorithm does not deteriorate with $N$. Thus, for large systems, MD can be more efficient than HMC, although there is room for methodological improvements.

## 5. Concluding remarks

Variational molecular dynamics (VMD) and variational path integral molecular dynamics (V-PIMD) methods have been developed. These meth-

ods are designed to accurately calculate ground state properties of many body systems using suitably introduced equations of motion. The VMD is a molecular dynamics analog of the variational Monte Carlo method. Square modulus of a trial wavefunction is reinterpreted to be a fictitious canonical distribution, then a molecular dynamics method for generating the canonical distribution is constructed. On the other hand, the V-PIMD is a molecular dynamics algorithm for a variational path integral method which can be used to numerically obtain an exact ground state. A molecular dynamics algorithm is constructed on the basis of the path integral molecular dynamics method developed for finite temperature systems. Two model systems have been tested as pilot calculations of the present methods. One is a one-dimensional harmonic oscillator as a simple model system that is analytically solvable. The VMD method perfectly works for various values of a variational parameter in the trial wavefunction employed and recovers the zero variance property of the total energy. On the other hand, the V-PIMD method is demonstrated to generate the exact ground state of the oscillator; total and potential energies and distribution function of the oscillator are found to be in good agreement with analytical results. The other model system is the liquid helium-4 as a realistic application. Total energy by the V-PIMD method agrees well with experimental value. Radial distribution function by the V-PIMD is found to be much closer to experimental one than that by the VMD method. It indicates that the description of the ground state is largely improved by the V-PIMD method, starting from a trial wavefunction used in the VMD calculation.

In the present study, the molecular dynamics methods have been applied

to the simple model systems. Methodological improvements are important for large scale quantum simulations. For example, in V-PIMD, higher order factorization schemes of the density matrix [23, 24, 25] may be useful to reduce the number of time slices. In the higher order schemes, however, we need derivatives of the interaction potential; the use of the higher order approximations is computationally more expensive than that of the primitive approximation (PA). It has been suggested [23] that using the molecular dynamics method, computational cost can be reduced close to that by PA using multiple time step algorithms, in which the higher order terms by the derivatives do not have to be evaluated at every time step. The hybrid Monte Carlo algorithm [10, 22], which can be easily constructed by the present MD method, also may provide an efficient way to perform variational path integral simulations with the higher order approximations; short MD runs to generate trial configurations are performed without the higher order terms, which are included in the Metropolis criterion. On the other hand, it is interesting to extend the variational molecular dynamics method for dynamically searching variational parameters' space. This could be realized by choosing the variational parameters to be additional dynamic variables and employing an adiabatic molecular dynamics technique developed by Rosso *et al.* [26]. These issues will be addressed in the near future.

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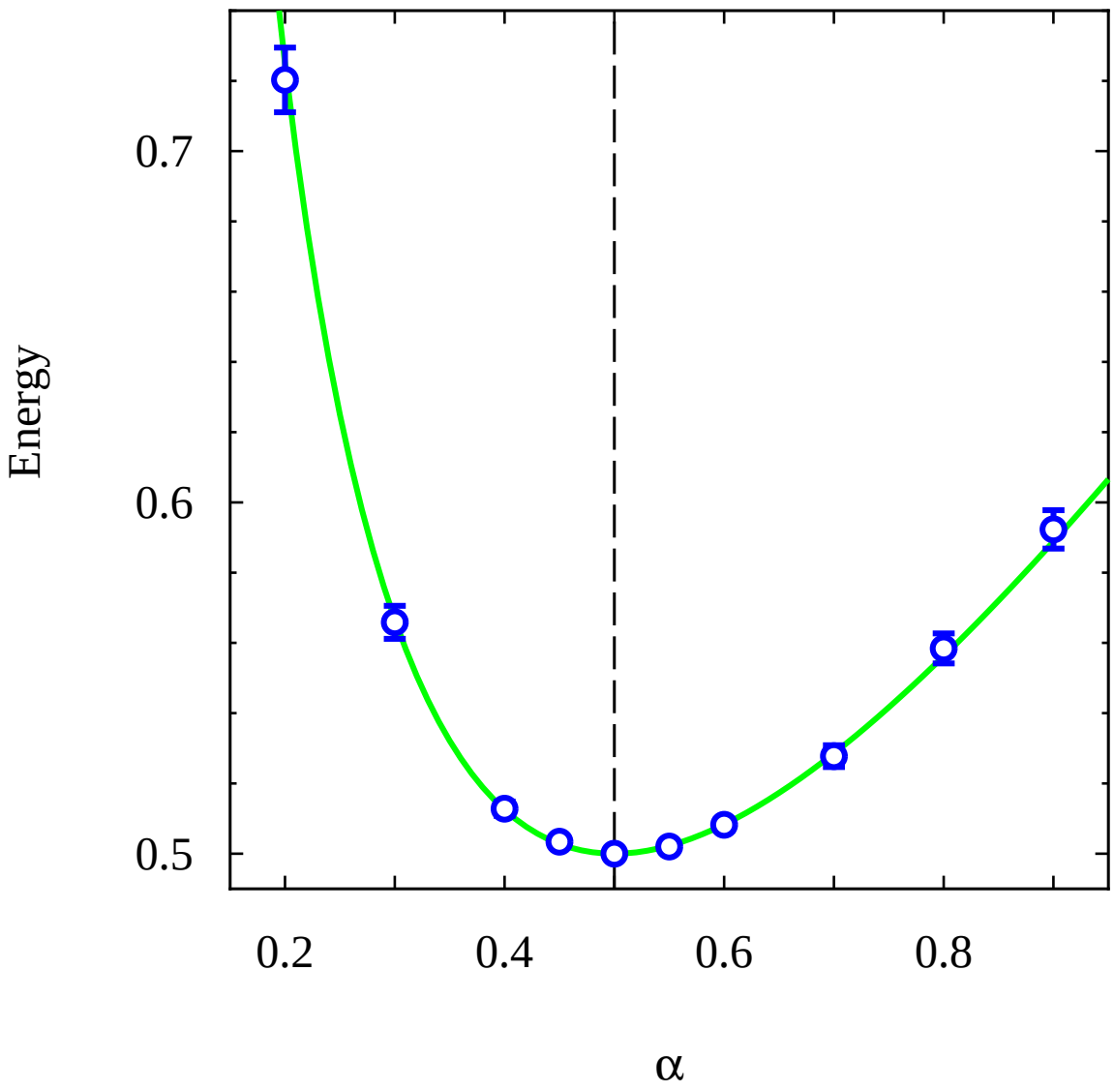


Figure 1: Total energy of the one-dimensional harmonic oscillator at the ground-state as a function of the variational parameter $\alpha$. Green solid curve indicates the analytical result of the total energy using the trial wavefunction, Eq. (13). Blue open circles are the variational molecular dynamics results. The error bar is expressed at 95 % confidence level, and is smaller than the size of the corresponding data symbol when it is not shown.

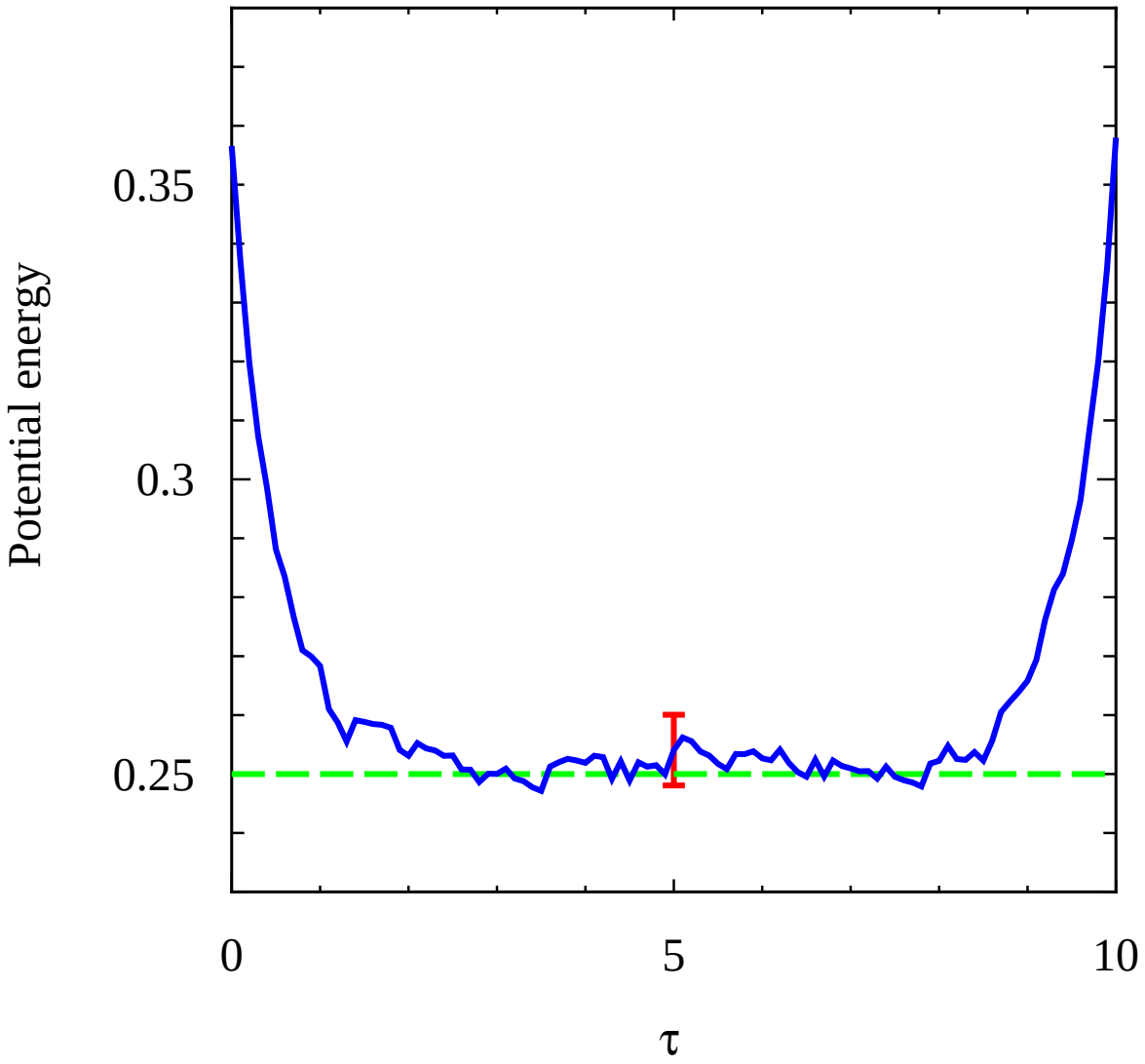


Figure 2: Averaged potential energy of the one-dimensional harmonic oscillator as a function of the imaginary time $\tau$. Blue solid line is for the variational path integral molecular dynamics results. Green dashed line indicates the exact quantum mechanical potential energy of the oscillator, 0.25. The error bar is expressed at 95 % confidence level, for the V-PIMD potential energy at $\tau = 5.0$.

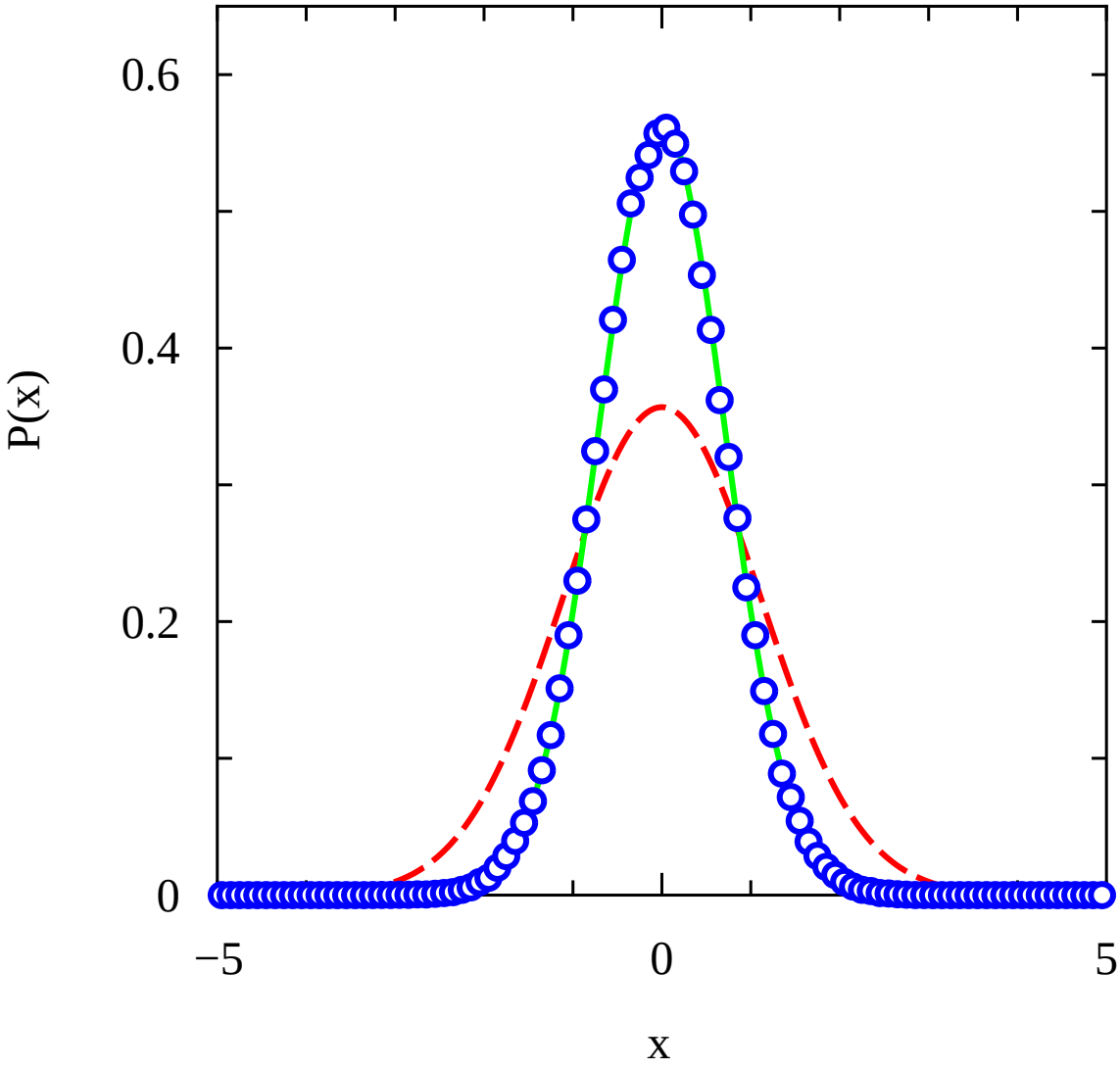


Figure 3: Spatial distribution of the one-dimensional harmonic oscillator as a function of the coordinate $x$, $P(x)$. Blue open circles are for variational path integral molecular dynamics results. Blue solid line is for the exact $P(x)$ of the oscillator and red dashed line for $P(x)$ using the trial wavefunction with the variational parameter $\alpha = 0.7$.

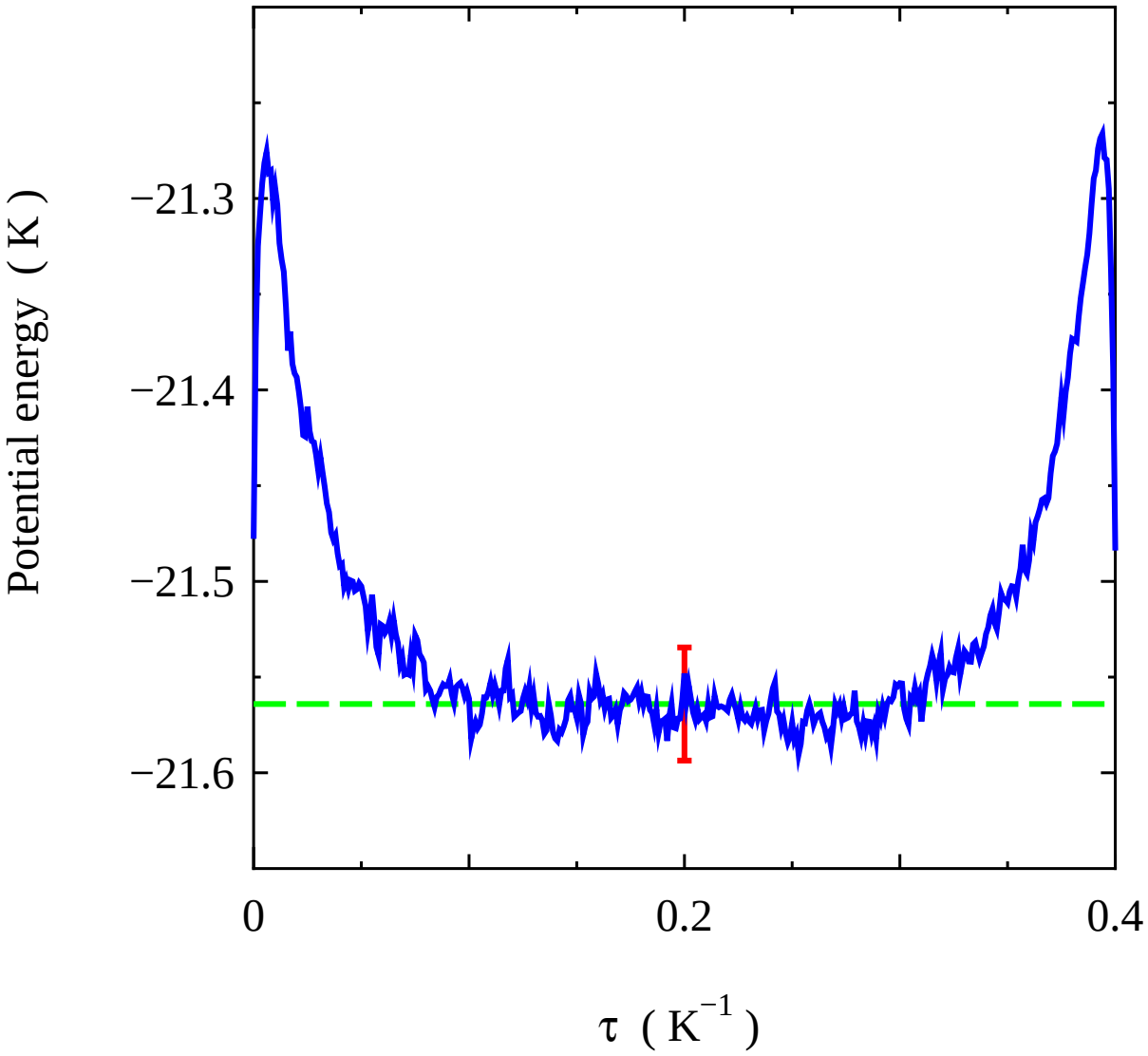


Figure 4: Averaged potential energy of the liquid helium-4 per atom as a function of the imaginary time $\tau$. Blue solid line is for the variational path integral molecular dynamics result and blue dashed line indicates the averaged potential energy at $\tau = 0.2$. The error bar is expressed at 95 % confidence level for the potential energy at $\tau = 0.2$.

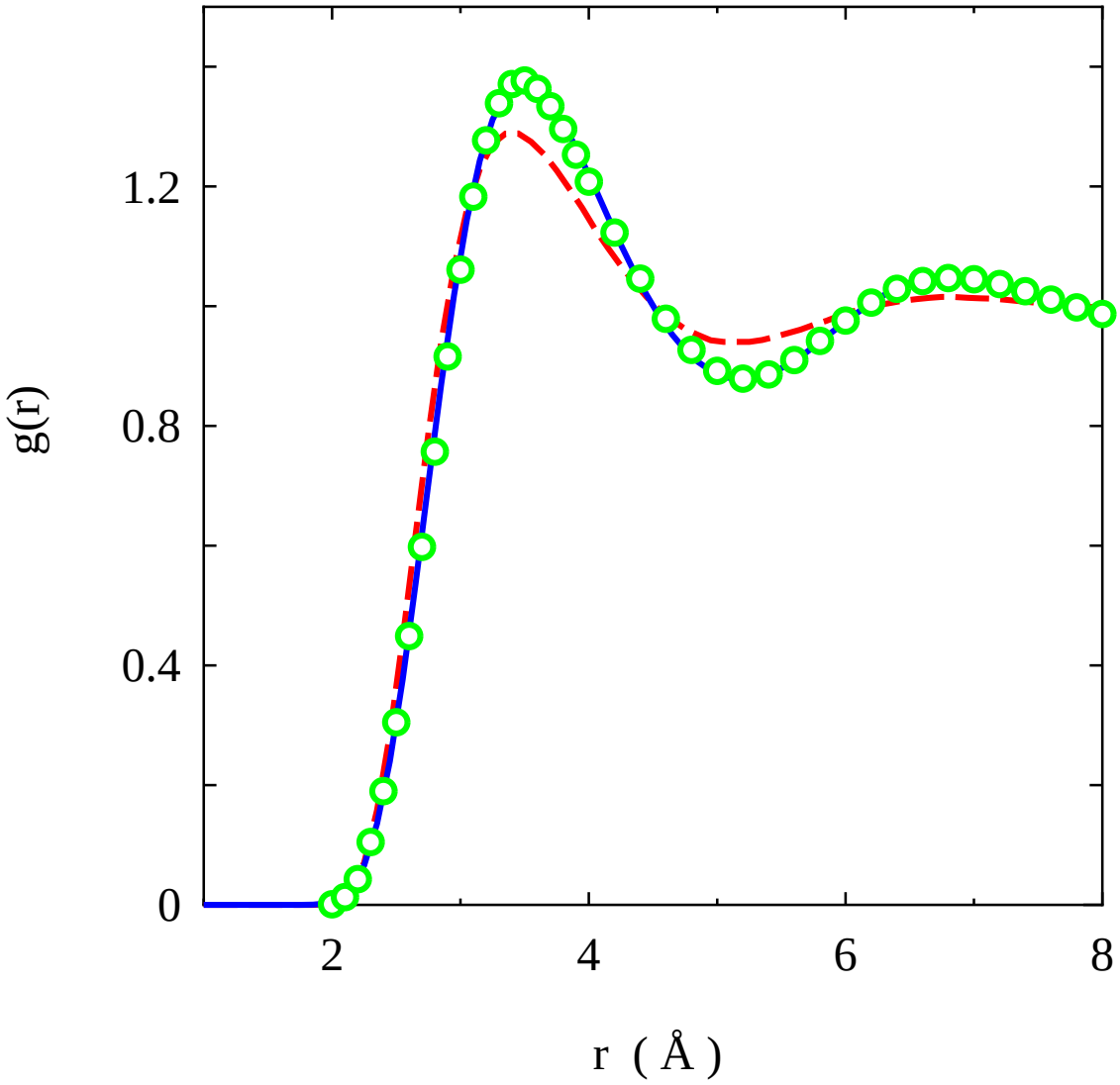


Figure 5: Radial distribution function of the liquid helium-4, $g(r)$. Blue solid line is for the variational path integral molecular dynamics result and red dashed line is for the variational molecular dynamics result with the variational parameter $b = 3.07$ Å. Green open circles are for the experimental results [17].

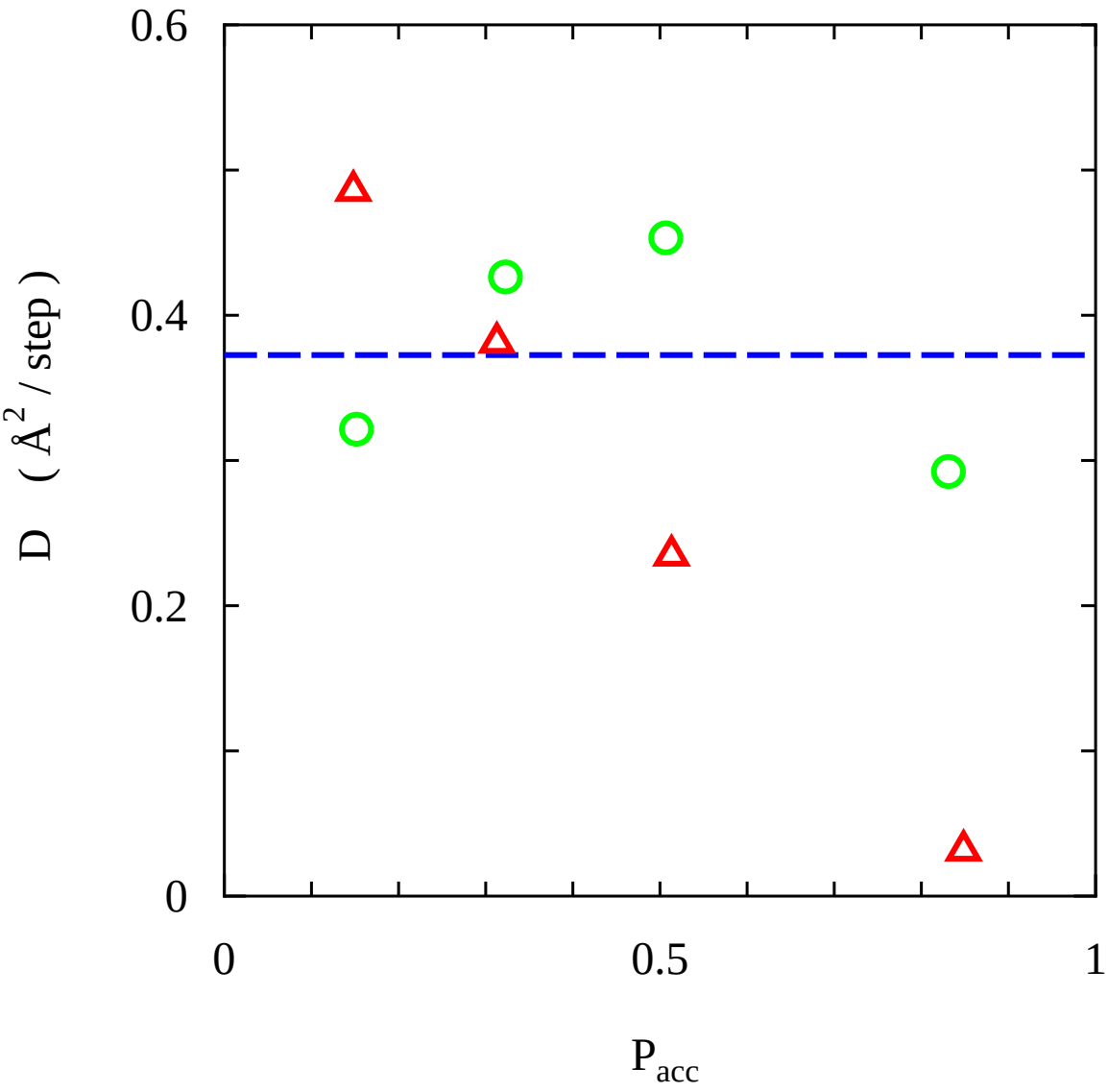


Figure 6: Self-diffusion coefficient $D$ as a function of the acceptance ratio $P_{acc}$ for the liquid helium, which is defined by the slope of the mean square displacement of a tagged atom as a function of MC or MD step. Red triangles are the results by the standard Metropolis Monte Carlo method and green circles are by the smart Monte Carlo method. Dashed blue line indicates the variational molecular dynamics result.